

PB88-125661

Isoprene Emission Fluxes
Determined by an Atmospheric
Tracer Technique

Washington State Univ., Pullman

Prepared for

Environmental Protection Agency
Research Triangle Park, NC

1986



U.S. Department of Commerce
National Technical Information Service

NTIS

TECHNICAL REPORT DATA		
(Please read Instructions on the reverse before completing)		
1. REPORT NO. EPA/600/J-86/412	2.	3. RI PB88-125661
4. TITLE AND SUBTITLE ISOPRENE EMISSION FLUXES DETERMINED BY AN ATMOSPHERIC TRACER TECHNIQUE (Journal Version)	5. REPORT DATE 1986	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) Brian Lamb, Hal Westberg, and Gene Allwine	8. PERFORMING ORGANIZATION REPORT NO.	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Laboratory for Atmospheric Research Washington State University Pullman, Washington 99164	10. PROGRAM ELEMENT NO. CDWA1A:02 - 1143 (FY-84)	11. CONTRACT/GRANT NO. CR 809296-02
12. SPONSORING AGENCY NAME AND ADDRESS Atmospheric Sciences Research Laboratory-RTP, NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, North Carolina 27711	13. TYPE OF REPORT AND PERIOD COVERED	
	14. SPONSORING AGENCY CODE EPA/600/09	
15. SUPPLEMENTARY NOTES Atmospheric Environment, 29(1):1-8, January 1986		
16. ABSTRACT The report describes a series of tracer release experiments to determine isoprene emission flux from an isolated oak forest area in Goldendale, Washington. Controlled releases of sulfur hexafluoride (SF₆) were performed within the forested area at 6 to 9 strategically located release points to simulate the release of isoprene from the forest vegetation. Samples for isoprene and SF₆ analyses were collected at 100 and 250 meter distances downwind of the forested area. Using biomass to surface area assumptions, emission rates were calculated. Emission rates determined with this technique were compared to measurements made by a bag enclosure procedure. The regression lines based on tracer data and enclosure results versus temperature have nearly identical slopes and the isoprene fluxes predicted from the two methods agree within 35% or less at all temperatures studied. The 840 to 31000 µg/m²/hr range of emission flux are comparable to measurements made in Tampa, Florida, Atlanta, Georgia, and Houston, Texas.		
17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
REPRODUCED BY U.S. DEPARTMENT OF COMMERCE NATIONAL TECHNICAL INFORMATION SERVICE SPRINGFIELD, VA. 22161		
18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS (This Report) UNCLASSIFIED	21. NO. OF PAGES 10
	20. SECURITY CLASS (This page) UNCLASSIFIED	22. PRICE PC12.95/mf 6.95

ISOPRENE EMISSION FLUXES DETERMINED BY AN ATMOSPHERIC TRACER TECHNIQUE

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(First received 18 March 1985 and in final form 20 May 1985)

Abstract—Sulfur hexafluoride tracer was used in a series of experiments to simulate isoprene emissions from an isolated oak grove. The measured tracer release rate and ambient concentrations of isoprene and SF_6 observed along downwind sample lines were combined to determine the mass flux of isoprene from the forest. The fluxes determined from the tracer data increased exponentially with temperature and were in close agreement with estimates determined from emission rates measured in a series of branch enclosure samples. The results of this field study demonstrate the feasibility and usefulness of simulating forest emissions as a tool for investigating turbulent transport in forested areas. Isoprene emission fluxes that can be applied in regional models are reported.

Key word index: Isoprene, tracer, SF_6 , natural emissions, biogenic.

INTRODUCTION

The mass fluxes of biogenic gases released from a forest have previously been determined using either micro-meteorological gradient methods (Knoerr and Mowry, 1981; Lamb *et al.*, 1985) or vegetation enclosure techniques (Zimmerman, 1981). Although these various approaches yield mass fluxes of similar magnitude, confidence in the results has been limited by uncertainties in the effects of enclosing vegetation, in measurements of small vertical gradients above the forest, and in the applicability of surface layer theory to nonideal sites. In many respects, the difficulties involved in measuring mass fluxes from a forest can be avoided by simulating the forest emissions with an inert tracer release and measuring ambient concentrations of the tracer and biogenic gases along downwind sample lines. The tracer approach does not require perturbation of the vegetation nor does it rely upon precise gradient measurements.

In an earlier paper (Allwine *et al.*, 1984), we describe the application of this technique to the simplest situation, an isolated forest. Different calculation methods were tested based upon the known tracer release rate and the measured ambient concentrations of the gases, and the results were compared to emission fluxes determined with a vegetation enclosure method. In this manuscript, additional tracer experiments are described which supplement the preliminary work described previously. Isoprene emission fluxes derived from tracer experiments are reported along with a discussion of various factors that affect the validity of the tracer methodology. This study provides isoprene emission flux estimates that can be used in regional

models. It also furnishes new insights concerning the relationship between biogenic emission rates and ambient concentrations of natural hydrocarbons.

EXPERIMENTAL PROCEDURES

For these initial studies, we selected the simplest emission source—an isolated grove of oak trees. Oaks emit significant quantities of isoprene which is a biogenic hydrocarbon that can be easily measured using gas chromatographic techniques. By selecting a small, isolated source, the upwind biogenic contribution was minimized and the biological and physical characteristics of the source region could be clearly established.

The study site was located approximately 40 km northeast of Goldendale, Washington, in the south central part of the state. The location is approximately 750 m above sea level with agricultural land on the north and east and range land on the west and south. The forest consists entirely of Oregon white oak (*Quercus garryana*). Undergrowth beneath the uniform and dense canopy was very sparse consisting mostly of range grass. Summertime meteorological conditions are typically dry and hot with prevailing winds from the southwest.

A total of 22 tracer experiments were conducted during the late summer and fall in 1982 and 1983. Seventeen of the tests provided adequate data for characterizing isoprene fluxes from the oak grove. Wind direction changes during five of the tests caused the isoprene and SF_6 plumes to miss the sampling area. During each test, sulfur hexafluoride (SF_6) was released for approximately 70 min at a constant rate. The total release rate was approximately 150 g h^{-1} . The SF_6 was contained in a single gas cylinder and released through a pressure regulator and rotameter to a manifold leading through 30 m lengths of tubing to either 6 or 9 release points within the forest canopy. A map of the grove with the release points is shown in Fig. 1. The release points were positioned approximately 2 m above the ground. The actual release rate from each point was measured with a bubble flowmeter several times during the release. The measured emission rates from each point are estimated to be accurate within $\pm 5\%$.

Two sample lines were layed out at distances of approximately 100 and 250 m downwind of the grove. Typically,

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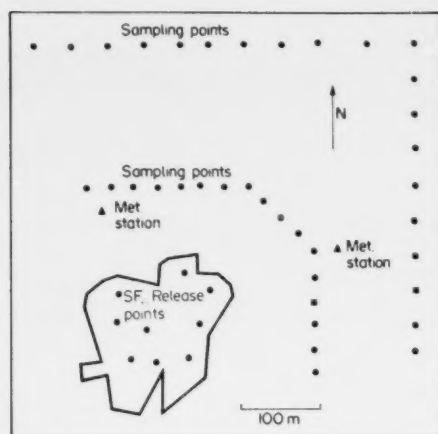


Fig. 1. Map showing location of oak grove, SF_6 release points and downwind sample lines.

10 tracer and 8 hydrocarbon samplers were located along a line. The sampling locations utilized during the various tests are shown in Fig. 1. Portable battery operated samplers (Krasnec *et al.*, 1984) were used to obtain four sequential 15-min average tracer samples in 20 cm^3 syringes. Whole air samples for hydrocarbon analyses were collected at selected points using battery operated Teflon pumps connected to 10 l Tedlar bags. Each pump and bag was placed in a box to shield them from sunlight. After each test, the contents of the bags were pumped into stainless steel canisters and returned to the laboratory.

Gas chromatographic analyses for the isoprene samples were performed with a Hewlett Packard chromatograph equipped with subambient temperature programming capabilities. A SE-30 glass capillary column (J & W Scientific) provided good resolution of isoprene. Helium was used as the carrier gas, with column flow maintained at 1 $\text{cm}^3 \text{min}^{-1}$. Optimum results were obtained by temperature programming from -50° to 80°C at 4°min^{-1} . Tracer samples were analyzed using custom-built electron capture gas chromatographs equipped with 5A 80/100 mesh molecular sieve columns ($8' \times 1/8''$ O.D.) and 1 cm^3 gas sampling loops. Additional details concerning the hydrocarbon and SF_6 analyses are available elsewhere (Allwine *et al.*, 1984).

RESULTS AND DISCUSSION

Preliminary results from the experiments conducted during 1982 were described in an earlier paper (Allwine *et al.*, 1984). In the initial tests, six SF_6 release points were utilized and it was assumed that this tracer release network accurately simulated the biogenic emission of isoprene. In addition, it was assumed that isoprene was conserved during the short transport distances from the source region to the sampling points. Two different methods were employed to calculate isoprene emission fluxes based on the known tracer release rate and measured ambient concentrations of isoprene and SF_6 . The simplest method involved only a consideration of the ratio of the observed maximum isoprene and SF_6 concentrations:

$$F_i = F_{\text{SF}_6} \times \frac{C_i(\text{max})}{C_{\text{SF}_6}(\text{max})} \quad (1)$$

where F_i is the calculated isoprene flux ($\mu\text{g m}^{-2} \text{h}^{-1}$), F_{SF_6} is the measured tracer flux ($\mu\text{g m}^{-2} \text{h}^{-1}$), $C_i(\text{max})$ is the maximum isoprene concentration and $C_{\text{SF}_6}(\text{max})$ is the maximum tracer concentration observed along the sample line.

In some cases, the horizontal distribution of SF_6 concentrations appeared to be slightly wider than and offset from the isoprene concentration distributions. For these situations, the calculations can be improved by considering the ratio of the crosswind integrals:

$$F_i = F_{\text{SF}_6} \frac{\int_{-\infty}^{\infty} C_i dy}{\int_{-\infty}^{\infty} C_{\text{SF}_6} dy} \quad (2)$$

where $C_i(y)$ is the crosswind distribution of the downwind isoprene concentrations and $C_{\text{SF}_6}(y)$ is the crosswind distribution of the SF_6 concentrations. In this approach, it is assumed that the vertical dispersion rates for both gases are equal.

The crosswind integrals of SF_6 and isoprene for use in Equation (2) were determined by direct integration of measured concentrations and also by integration of the best-fit Gaussian curve through the measured observations. In both instances, the position of the sample points were projected onto the line normal to the mean wind direction, and the concentration profiles were integrated using trapezoidal integration. In almost all tests, the observed concentration profiles were Gaussian in appearance, but in several instances the measured profile did not encompass the entire SF_6 or isoprene plume. By employing best-fit curves to the data, the profile of the entire plume could be obtained, and integration errors associated with chopping off a portion of the plume were eliminated. The approach for calculating best-fit Gaussian curves was initially developed to perform tracer mass balances in tracer dispersion studies and is described in detail elsewhere (Lamb, 1978; Sackinger *et al.*, 1982). The use of best-fit Gaussian curves does not imply application of the common Gaussian plume model; rather the use of the curves only takes advantage of the Gaussian shape of the observed concentration profiles.

Another method for estimating isoprene emission rates from a forest is to calibrate an atmospheric dispersion model for the existing conditions using the SF_6 data and then apply the model to isoprene transport to determine the best estimate of the emission flux. Arnts *et al.* (1982) employed the Point Area, Line Source (PAL) Gaussian algorithm to estimate alpha-pinene flux from a loblolly pine forest. Tracer data were used to define the best Pasquill-Gifford dispersion curves for the test conditions. However, application of a simple Gaussian plume model in the present case does not appear to provide any unique or improved results, and this modeling approach will not be tested with the data from the isolated forest.

Table 1 provides a summary of experimental conditions for each test and lists the isoprene fluxes calculated by each of the above methods. The fluxes determined from the ratio of the maximum concentra-

Table 1. Summary of test conditions and calculated isoprene fluxes

Test	Date	Start time	WS (m s^{-1})	WD (deg)	Temp* (°C)	SF ₆ flux ($\mu\text{g m}^{-2} \text{h}^{-1}$)	C _i -Back† ($\mu\text{g m}^{-3}$)	C _i -max‡ ($\mu\text{g m}^{-3}$)	C _{SF₆} -max ($\mu\text{g m}^{-3}$)	Isoprene flux§ ($\mu\text{g m}^{-2} \text{h}^{-1}$)	
										Method 1	Method 2
1	16/8/82	1530	5.5	270	26	20300	0.8	3.9	4.3	18400	17100
2	17/8/82	1000	1.0	194	24	20600	0.7	1.5	2.5	12180	10500
3	31/8/82	1600	3.0	217	27	17500	1.0	2.0	4.5	7780	11500
4	31/8/82	1745	3.6	241	23	17500	0.9	1.4	6.8	3660	4470
5	8/9/82	1300	5.8	247	27	20300	1.7	4.6	3.5	26600	31000
6	8/9/82	1500	6.2	240	28	20300	1.7	6.5	6.2	21000	27100
7	22/9/82	1300	2.2	145	21	19600	0.2	1.4	7.2	3800	4060
8	22/9/82	1500	3.1	118	22	19600	0.3	1.0	4.2	4660	5760
9	28/9/82	1330	8.5	260	11	20800	0.1	0.3	5.2	1200	840
10	19/8/83	1300	1.5	210	21	24000	2.0	3.6	6.3	13800	15800
11	19/8/83	1530	2.8	210	20	24300	1.3	1.1	2.6	10300	8340
12	27/8/83	1100	1.1	225	20	28500	1.1	0.9	2.1	12800	9500
13	27/8/83	1400	1.3	230	24	25600	1.1	2.7	3.5	26800	20700
14	8/9/83	1130	1.6	250	15	21900	1.2	2.4	9.3	5700	3580
15	8/9/83	1400	1.8	250	16	21600	0.8	1.5	13.9	2330	1530
16	16/9/83	1130	2.2	269	18	22700	1.0	2.6	5.2	11400	7370
17	16/9/83	1400	3.2	246	18	22100	1.0	1.1	4.8	5060	840

*Ambient air temperature.

†Upwind isoprene concentrations.

‡Maximum downwind isoprene concentrations minus upwind isoprene concentrations.

§Method 1 flux based on ratio of maximum concentrations; Method 2 flux based on ratio of crosswind integrals.

tions (Method 1) range from 1200 to $26800 \mu\text{g m}^{-2} \text{h}^{-1}$ with a geometric mean flux of $8110 \mu\text{g m}^{-2} \text{h}^{-1}$. Fluxes calculated with the ratio of the observed crosswind concentration integrals (Method 2) vary from 840 to $31000 \mu\text{g m}^{-2} \text{h}^{-1}$ with a geometric mean flux of $6780 \mu\text{g m}^{-2} \text{h}^{-1}$. The mean temperature for each method was 21°C .

The strong dependence of isoprene emission rates upon temperature has been well documented (Flyckt, 1979; Tingey *et al.*, 1981; Lamb *et al.*, 1985). As a result, the degree of correlation between the estimated fluxes and temperature can be used as a basis of comparison of the two calculation methods. As shown in Fig. 2, isoprene fluxes calculated by both procedures exhibit a positive exponential relationship with ambient temperature. The correlation coefficient for the calculations based upon the ratio of the maximum concentrations was 0.74, which is slightly less than the correlation coefficient based upon the ratio of the integrals (0.83). It can be seen in Fig. 2 that the regression lines intersect at about 24°C . Thus, emission fluxes calculated by the two methods are nearly

identical for mid-twenty degree temperatures. Method 1 yields an isoprene flux approximately 25% greater than Method 2 at 20°C , while at 28°C the difference is about the same but Method 1 now gives the lower flux. It appears that either method can be used to calculate emission fluxes; however, since the crosswind integral procedure (Method 2) yields a better correlation coefficient and involves a more rigorous treatment of the data, it is probably the better of the two approaches.

In order to test the validity of the tracer methodology, direct measurements of isoprene emission rates were made using the enclosure technique (Zimmerman, 1981). Thirteen enclosure measurements were made concurrently with the 1982 tracer experiments. Table 2 lists enclosure sampling conditions and calculated isoprene emission rates.

In order to convert the emission rates listed in Table 2 to an emission flux, a foliage biomass factor (g m^{-2}) had to be determined for the forest. A forest inventory was conducted to determine the average height of the forest (8 m), average number of trees per hectare

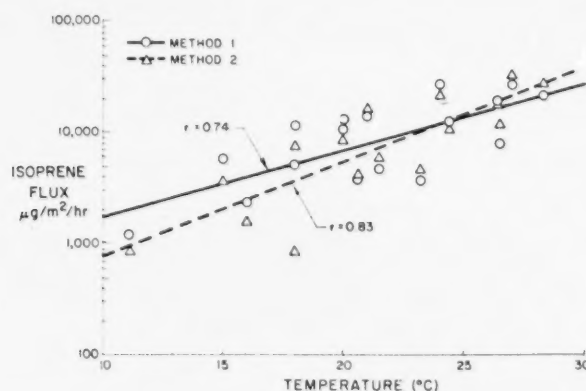


Fig. 2. Isoprene flux vs ambient temperature derived from the ratio of the observed maximum isoprene and SF_6 concentrations (Method 1) and from the ratio of the observed isoprene and SF_6 crosswind integrals (Method 2).

Table 2. Isoprene emission rates from branch enclosure samples

Sample	Time (PST)	Date 1982	Ambient temperature ($^\circ\text{C}$)	Enclosure temperature ($^\circ\text{C}$)	Emission rate ($\mu\text{g g}^{-1} \text{h}^{-1}$)
1	1220	17/8	27.2	33	130
2	1410	31/8	25.0	24	94
3	1818	31/8	23.9	22	0.9
4	0843	8/9	19.0	19	1.1
5	1309	8/9	27.7	29	36
6	1525	8/9	28.3	33	110
7	0924	14/9	10.0	11	0.4
8	1235	14/9	13.3	15	4.0
9	1451	14/9	14.4	18	8.0
10	1034	22/9	16.7	19	0.8
11	1524	22/9	21.7	24	2.6
12	1544	22/9	21.7	22	1.2
13	1343	28/9	11.1	16	7.3

(2800), the average tree diameter at breast height (DBH) (14.2 cm), and the total area of the grove (1.5 ha). Results from this inventory were used with empirical relationships reported for a variety of deciduous species (Monk *et al.*, 1970; Reiners, 1972; Sollins *et al.*, 1973; Whittaker *et al.*, 1974) to yield dry weight foliage biomass factors in the range $574\text{--}1030\text{ g m}^{-2}$. This range is somewhat higher than deciduous biomass factors estimated for other sites. For example, Zimmerman (1981) used 450 g m^{-2} as a national average. The higher range estimated for the oak grove is attributed primarily to the large number of trees per unit area.

By applying a biomass factor of 804 g m^{-2} which is the mean of the range listed previously, a relationship between temperature and emission flux was derived as shown in Fig. 3. The regression lines based on tracer data (Method 2) and enclosure experiments have nearly identical slopes and the isoprene fluxes predicted from the two methods agree extremely well at all temperatures. The excellent agreement between the slopes of the lines from the two independent methods indicates that the emission flux measurements from both procedures are reasonable estimates. Although the actual biomass factor is uncertain, the range of reported factors yields flux estimates that bracket the flux determined from the tracer experiments. The close agreement in emission fluxes when a midpoint biomass factor of 804 g m^{-2} is applied suggests that this is a reasonable factor to use for the oak grove employed in this study.

Previously it was pointed out that two assumptions had been made in conjunction with the tracer experiments, namely: (1) that the trace release network with six emission points within the grove provided an accurate simulation of the isoprene emissions, and (2) that isoprene was conserved (i.e. no chemical loss) between the emission source and downwind sample points. In addition to providing more data for establishing isoprene fluxes, the 1983 field experiments

were designed to substantiate these assumptions. Three additional tracer release points were incorporated in 1983 and the sampling network was enlarged through addition of a second sample line 150 m further downwind. If the original six tracer release points had adequately simulated the isoprene emissions, the three additional release points added in 1983 should not change the original flux estimates. Incorporation of the second sample line at a greater distance from the source provided a means of determining whether or not isoprene was conserved during transport. Evidence for chemical losses would exist if fluxes calculated from data collected along the most distant line were significantly less than those derived from the inner sampling line.

Figure 4 provides a comparison of the isoprene flux-temperature relationships obtained with six and nine tracer release points. As can be seen, isoprene fluxes are very similar for temperatures in the $10\text{--}15^\circ\text{C}$ range but differ by more than a factor of two at temperatures above 22°C . At present, we do not know whether this difference is a function of the tracer release network or other unknown factors. Currently, little is known about the dependence of biogenic emissions on environmental parameters other than temperature. It is apparent that additional tracer experiments will be required in order to resolve this anomaly. A series of tests in which the tracer release network is changed over a short time frame is needed.

The assumption that isoprene was conserved during transport is reasonable because of the short travel distances between the source and sample lines. Chemical lifetimes for isoprene range from 1.3 to 34 h depending on the ambient ozone and hydroxyl concentrations (Altshuler, 1983). For conditions utilized in these tracer tests, the maximum transport time from the oak grove to the most distant sample line (250 m) was less than 0.1 h. Using even the shortest lifetime for isoprene estimated by Altshuler (corresponding to 80 ppb O_3 and $10^{-7}\text{ ppm OH}^\cdot$), the loss of isoprene

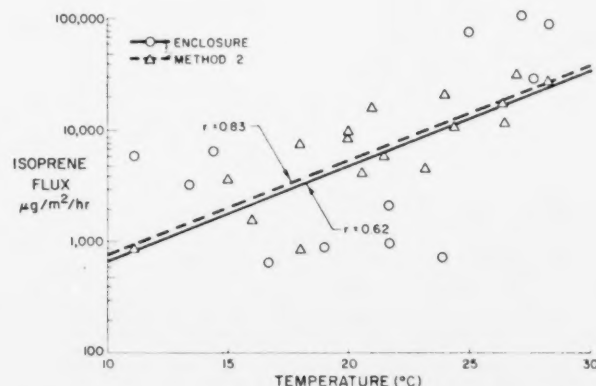


Fig. 3. Comparison of isoprene emission flux vs temperature determined from the tracer and enclosure procedures.

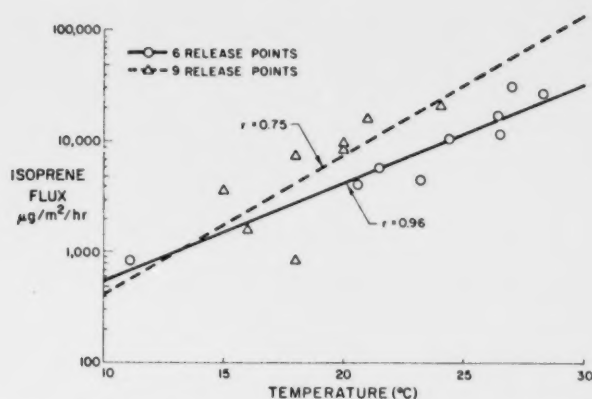


Fig. 4. Comparison of isoprene fluxes derived from tracer experiments employing six and nine release points within the oak grove.

during transport from the forest to the furthest sample line is estimated to be less than 8%. Flux estimates derived from experiments in which sample lines at downwind distances of 100 and 250 m were employed showed no evidence of chemical loss. For example, an isoprene flux of $20700 \mu\text{g m}^{-2} \text{h}^{-1}$ was determined from data collected along the 100 m sample line on the afternoon of 27/8/83. Further downwind along the 250 m line, an emission flux of $17700 \mu\text{g m}^{-2} \text{h}^{-1}$ was calculated. This difference of 15% in isoprene flux between the two sample lines is well within the experimental uncertainty associated with the tracer methodology. Even if a portion of this difference could be ascribed to chemical reaction, the isoprene loss between the source and the 100 m sample line would be less than 2% even for the shortest estimated lifetime

for isoprene. Data obtained along the 100 m sample line were used to derive the emission fluxes reported in Table 1.

The range and magnitude of the isoprene fluxes determined in this work are quite similar to fluxes obtained in other locations as shown in Table 3. At the same time, the ambient concentrations of isoprene measured downwind of the isolated oak forest are comparable to ambient isoprene concentrations reported for other locations. For example, in the present work, the emission fluxes varied from 840 to $31000 \mu\text{g m}^{-2} \text{h}^{-1}$ and the maximum ambient concentrations measured downwind of the oak grove were between 0.3 and $6.5 \mu\text{g m}^{-3}$. As the tracer data established, this concentration range is the result of atmospheric dilution of isoprene emitted from a

Table 3. Comparison of isoprene emission rates and ambient concentrations from various field and laboratory studies

Investigator	Species sample	Sample method	Isoprene emission Rate 30°C ($\mu\text{g g}^{-1} \text{h}^{-1}$)	Flux* ($\mu\text{g m}^{-2} \text{h}^{-1}$)	Ambient concentrations ($\mu\text{g m}^{-3}$)
Flyckt (1979)	Red oak, Pullman Washington	Branch enclosure Method	38.9	31300	—
Lamb <i>et al.</i> (1985)	5 hardwood species, Pennsylvania	Branch enclosure Method	27.5	22100	—
Lamb <i>et al.</i> (1985)	Oak-hickory forest, Pennsylvania	Aerodynamic Method	30.7	24700	3.0–15.0
Lamb <i>et al.</i> (1985)	Mixed deciduous species	Branch enclosure	22.4	18000	2.7 (ave)
Goldendale Study	Oregon white oak, Goldendale, WA	Tracer Method	40.5	32600	0.3–6.5
Goldendale Study	Oregon white oak, Goldendale, WA	Branch enclosure Method	42.7	34300	—
Zimmerman (1977)	Oak species	Branch enclosure Method	24.7	19900	~2.5
Tingey <i>et al.</i> (1978)	Live oak	Laboratory growth Chamber	33.0	26500	—

*Flux calculated using a biomass factor of 804 g m^{-2} .

diffuse source. Because of the short transport times and clean, rural conditions at the test site, loss of isoprene through chemical reaction was not an important factor in the observed concentrations. In the Pennsylvania work by Lamb *et al.* (1985), isoprene concentrations in the range of $3\text{--}15\ \mu\text{g m}^{-3}$ were observed 6 m above the canopy and emission fluxes from 2500 to $16000\ \mu\text{g m}^{-2}\text{ h}^{-1}$ were determined. Zimmerman (1979) measured an emission rate for one live oak branch of $29\ \mu\text{g g}^{-1}\text{ h}^{-1}$. He collected an ambient air sample close to the branch which contained an isoprene concentration of $2.5\ \mu\text{g m}^{-3}$. Westberg and Lamb (1983) reported nonmethane hydrocarbon (NMHC) concentrations at four sites in and around Atlanta, Georgia. They found a mean NMHC concentration of 476 ppbC of which 1% was determined to be isoprene ($2.7\ \mu\text{g m}^{-3}$). The estimated isoprene emission rate at 30°C from mixed deciduous species in Atlanta was $22\ \mu\text{g g}^{-1}\text{ h}^{-1}$. Cronn and Harsch (1979) collected air samples within the Great Smoky Mountain National Park in Tennessee. They reported an average ambient isoprene concentration of $3.4\ \mu\text{g m}^{-3}$ and a maximum concentration of $10\ \mu\text{g m}^{-3}$.

Since the levels of isoprene measured at Goldendale were entirely associated with dilution of the forest emissions, the reasonable agreement of these concentrations with those observed elsewhere suggest that atmospheric dilution of gases released from very diffuse sources can have a dominant effect upon the resulting ambient concentrations. The results presented here demonstrate that low ambient air concentrations of biogenic gases do not necessarily imply low source strengths or high chemical reactivity as has been suggested in previous discussions of the role of natural organics (Altshuller, 1983).

SUMMARY AND CONCLUSIONS

The simulation of isoprene emissions from an isolated grove of Oregon white oak with a multi-point SF_6 tracer release provided a straightforward method for determining the isoprene emission flux. Isoprene emission fluxes calculated from the measured tracer release rate and the ratio of the crosswind integrals of isoprene and SF_6 concentrations agreed closely with measurements of isoprene emission fluxes obtained with a vegetation enclosure technique. The fluxes determined with the enclosure procedure were based upon a biomass factor at the midpoint of a range of four available biomass algorithms reported for other deciduous forests. Emission rates from this work were also similar in magnitude to those reported for other isoprene-emitting hardwoods.

The range of ambient isoprene concentrations recorded during this study is essentially the same as reported by a number of other investigators for a variety of locations. The isoprene and tracer data together underscore the importance of atmospheric

dilution in the production of low ambient concentrations of biogenic hydrocarbons emitted from forested areas.

Acknowledgments—This work was supported by the U.S. Environmental Protection Agency (CR809206010). Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

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